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In Situ Laser Crystallization of Amorphous Silicon for TFT Applications: Controlled Ultrafast Studies in the Dynamic TEM

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In Situ Laser Crystallization of Amorphous Silicon for TFT Applications: Controlled Ultrafast Studies in the Dynamic TEM

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Abstract

An in situ method for studying the role of laser energy on the microstructural evolution of polycrystalline Si is presented. By monitoring both laser energy and microstructural evolution simultaneously in the dynamic transmission electron microscope, information on grain size and defect concentration can be correlated directly with processing conditions. This proof of principle study provides fundamental scientific information on the crystallization process that has technological importance for the development of thin film transistors.

Polycrystalline Si (poly-Si) is of technological interest for use in thin film transistors (TFT) for flat panel displays in rapidly growing areas, such as monitors, mobile phones and televisions [1-7]. Unfortunately, poly-Si produced for use in TFTs contains grain boundaries and intergranular defects, and the overall properties of poly-Si are dominated by the electrical activity of charge-trapping centers that they induce [4]. Many studies have shown that larger grain size results in a beneficial higher carrier mobility [1,4,5]. However, larger grain size does not totally mitigate the defect problem, as grain quality (defect, twin, and stacking fault concentration) and grain distribution also affect the overall device characteristics [4]. Therefore, it is necessary to understand the origin and role of inter and intragranular defects to control poly-Si microstructures for TFT applications.

Typically, industrial processing of poly-Si films proceeds through crystallization of amorphous Si (a-Si) with continuous, diode-pumped solid state, or excimer lasers Though ex-situ studies have been performed to understand the role of grain size and defects on device performance, it is imperative that a method be developed to study the development of microstructures *during* processing. We present a new method of processing a-Si films by in situ laser crystallization in a transmission electron microscope (TEM) with lasers fed into the electron column. In our experiments, we base our laser treatments on an industrial excimer laser processing method with pulse durations in the order of 50ns at an energy density of 330mJ/cm^2 for a 50nm film. Crystallization velocities for this type of experiment were previously found to be 10m/s [5]. This setup is called the Dynamic TEM, or DTEM, and is described in greater detail in [8-13]. The utility of the DTEM has been demonstrated for ultrafast in situ TEM experiments [8-11]

and unique laser processing of materials [12,13]. In this paper, we show the combined use of the laser processing with ultrafast imaging to study the crystallization of a-Si.

The DTEM consists of a modified TEM column (JEOL 2000FX) to which two pulsed lasers have been added: the "drive laser" and the "cathode laser". When using the DTEM, the drive laser is used to treat the sample directly (to initiate a phase change or reaction), while the cathode laser is used to treat the photoelectron cathode to emit electrons for pulsed electron beam imaging. A time delay set between these two laser pulses dictates the "pump-probe" experimental style, whereby the sample is pulsed first with the drive laser, and at a specific time after this pulse, the cathode laser hits the photoelectron cathode source to emit electrons to image the sample, finally caught on a CCD camera. The drive laser, a 8 ns duration Nd:YAG laser, can be operated at 1064, 532 or 355nm, and is incident directly on the sample (at approximately 43°) with an approximately 50μm diameter spot size. The laser energy can be adjusted for melting, heating, or ablation. The stimulated reaction by the drive laser energy is therefore investigated in situ during the experiment. The cathode laser, a 15-ns-duration frequency-quintupled Nd:YLF laser, enables operation in both a pulsed photoemission mode and in a conventional thermionic continuous wave (CW) mode. The pulsed mode's nanosecond-scale imaging capability greatly exceeds the ~30 Hz time resolution of conventional in situ TEM and has the potential to great insight into otherwise unknown aspects of microstructural evolution in various materials, especially during an amorphous to crystalline transition. We first present results obtained using conventional CW imaging mode, but taking advantage of the DTEM's in situ laser drive capability. Our studies in CW mode showed the success of attempting this unique, in situ, pulsed laser processing method of poly-Si films via crystallization of a-Si in a TEM. We then move on to taking full advantage of the DTEM's capabilities to study the microstructural evolution during laser processing on nanosecond time scales.

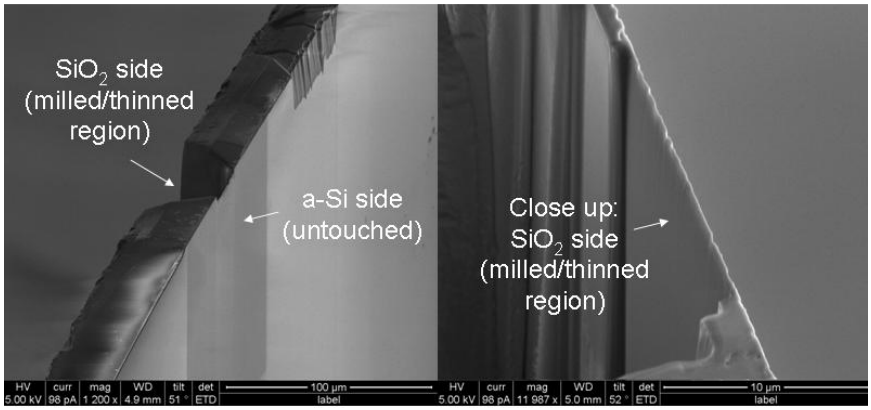

Figure 1. Secondary electron beam images showing FIB-thinned samples. Flat, untouched region shown on the left is the a-Si surface. Back-thinning process is shown on the right.

Wafers of 50nm a-Si/700nm SiO_2/glass, provided by Palo Alto Research Center (PARC), were made into electron transparent films using a FEI Nova600 Focused Ion Beam (FIB). First, the sample was ground from the SiO_2/glass side to a thickness of ~20μm. Then, the FIB was used prepare flat films of 50nm a-Si on ~150nm SiO_2 (as

opposed to conventional ion milling) by preferential thinning of the SiO_2/glass side. Initial ion-beam thinning was performed at 30kv and 20nA (only on the SiO_2/glass side) to a thickness of ~2μm. The sample was then subject to further ion beam thinning at progressively lower beam currents, from 0.3nA to 0.1nA, for an even distribution of 50nm a-Si/150nm SiO_2 film (**figure 1**). This sample geometry provides an accurate mimic to the excimer laser experiments performed in industry. Though films processed by excimer laser contain a glass backing, it couldn't be maintained in this case, as our samples needed to be thin enough for electron transparency in the TEM. We found, however, that 150nm of SiO_2 served as a sufficient cooling layer for crystallization to proceed. The film was processed in situ, inside the DTEM using the drive laser.

In our first experiments, we used conventional (thermionic emission) electron beam imaging to monitor nucleation and grain growth during laser crystallization by the frequency tripled Nd:YAG drive laser at 355nm. **Figure 2A and 2B** shows the sample after a laser shot at ~108 μJ and ~175.5 μJ, respectively, in a 50 μm diameter spot on the sample (an energy density of >108μJ/1963μm^2), revealing nucleation and growth of 50-100nm grains at lower energy, and >500nm grains at higher energy, from the melt. The films crystallized upon supercooling (nucleation and growth after the melt). In these experiments, we treated the film with multiple laser energies to watch the effect on the grain size. Much like in the industrial excimer laser experiments, the maximum grain size scaled with laser energy (**figure 2C**). However, we see in situ that the grain size distribution varies with Gaussian profile of laser energy density, similar to the results found in [14]. The largest grains are found directly around the center of the location where the laser hit the sample, and grain size trails off radially into smaller grains from this spot. Thus, we can predict the grain size distribution according to laser energy density.

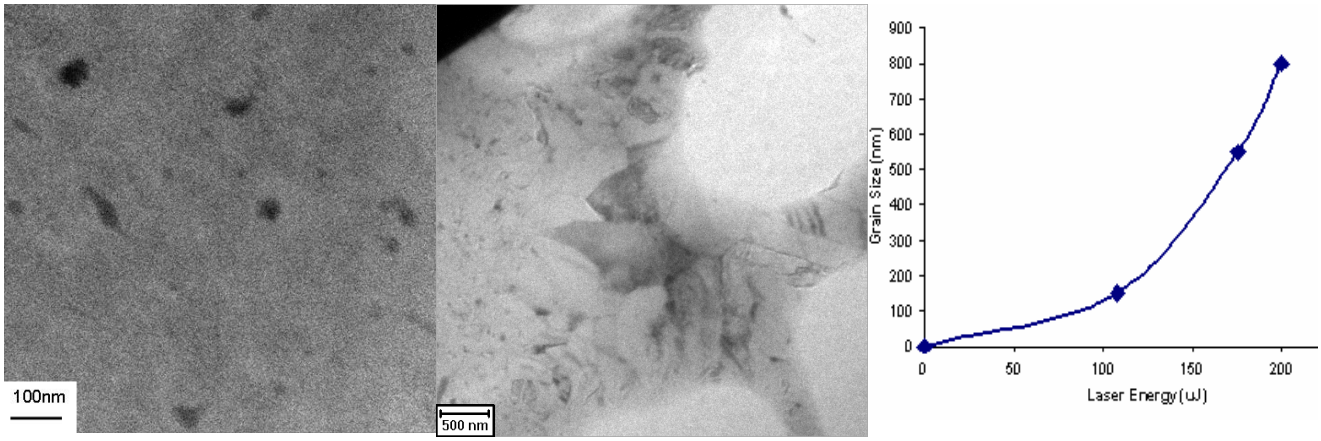

Figure 2. Thermionic (continuous wave) electron beam images showing plan views of film shot at ~108μJ (A) and at 175.5 μJ (B). Maximum grain size after crystallization experiment as a function of laser energy. Grain Size = approximate diameter of largest grain shown in field of view (C).

Though we successfully crystallized the a-Si films, looking at the grain image after hitting the film with a single shot of the drive laser in conventional CW imaging mode gave us little information about the origin and forming mechanism of defects and other microstructural characteristics. CW imaging is much slower than the DTEM's

pump-probe (drive and cathode laser) technique. Thus, we took advantage of the cathode laser in the DTEM for pulsed electron beam imaging, which allowed us to monitor growth processes that occur in a single drive laser shot with nanosecond time resolution. **Figure 3** shows the time resolved images of nucleation and growth front, in the bottom left corner of the middle and right images.

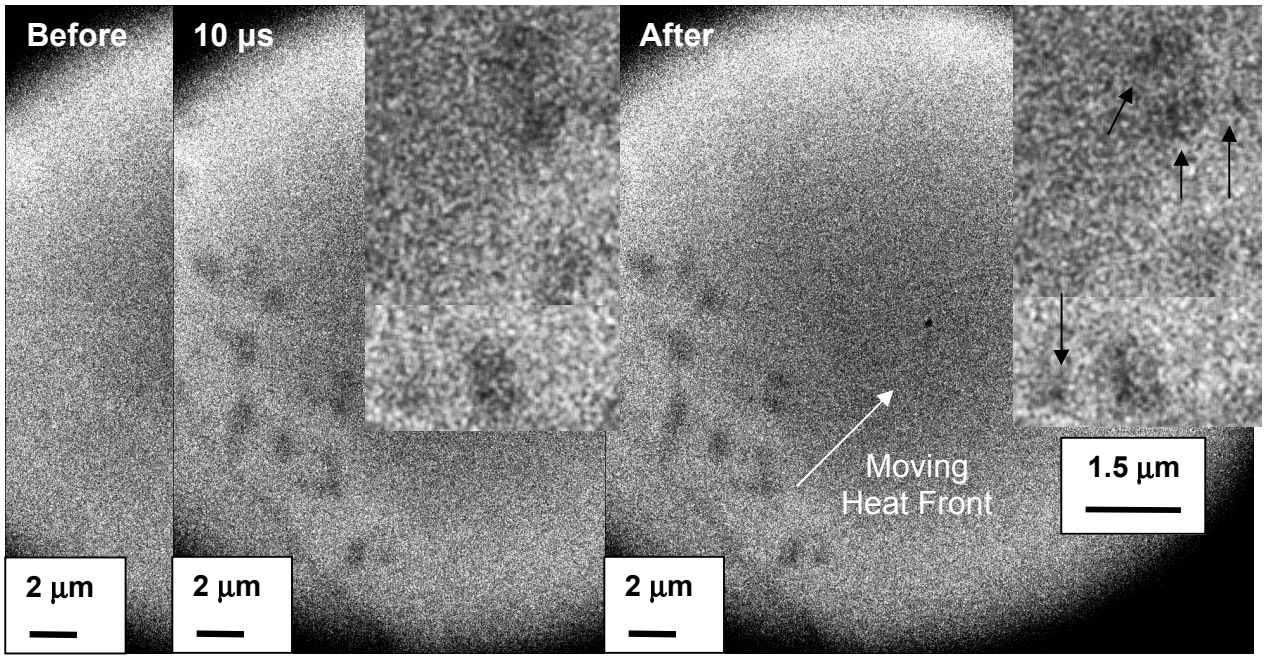

Figure 3. Time resolved, pulsed electron beam images of in situ laser crystallization. Nucleation and growth are occurring at the bottom left side of each image, where the heat front from the laser beam is located. Insets show pixilated close-up of 10 microsecond (left) and final (right) images. Circles (arrows) indicate two examples of where microstructural changes are seen between each image. Specifically, the grains that have become larger and darker in contrast indicate grain growth.

Small changes in the nucleation and growth of crystalline grains are seen by using time resolved imaging of the laser processing on nanosecond scales. In these images, we kept the magnification large to look at changes in multiple grains along the crystallization front. To the casual observer, the images look very similar. However, a closer look reveals changes in grain size and shape during the growth process. Specifically, growing and shrinking events during nucleation and growth are seen in the snapshots. To aid the eye, more pixilated views of the image sequences are shown the insets, with arrows around some examples of these changes during supercooling. These delicate microstructural changes were not seen using conventional thermionic imaging sequence (inset). The ability to view these kinds of transformations during crystallization is critical to the understanding of dominating microstructural features in laser processed poly-Si, how they behave, and how to control them. Small changes in grain size and shape during nucleation and growth can affect overall microstructure. Using this technique, we can finally understand intricate aspects of microstructural development as a function of laser energy. During crystallization, events such as defect formation can dictate overall electron mobility in the final microstructure. Using in situ laser crystallization, we can characterize when events like defect formation occur. Additionally, because the speed of

dislocations is too fast to be monitored by conventional video frame rate imaging, ultrafast imaging during laser crystallization can be used to pinpoint exactly what gives rise to these microstructural developments as a function of laser energy. Thus, with the combination of laser processing *inside* the TEM column, and time resolved in situ studies using nanosecond resolution, we are one step closer to understanding the intricate microstructural changes during laser processing of Si for TFT applications.

In conclusion, we successfully developed a method for studying UV laser processing of Si films in situ on nanosecond time scales, with ultimate implications for TFT application improvements. In addition to grain size distribution as a function of laser energy density, we found that grain size scaled with laser energy in general. We showed that nanosecond time resolution allowed us to see the nucleation and growth front during processing, which will help further the understanding of microstructural evolution of poly-Si films for electronic applications. Future studies, coupled with high resolution TEM, will be performed to study grain boundary migration, intergranular defects, and grain size distribution with respect to laser energy and adsorption depth.

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